Aerosol organic carbon to black carbon ratios: Analysis of published data and implications for climate forcing

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[1] Measurements of organic carbon (OC) and black carbon (BC) concentrations over a variety of locations worldwide have been analyzed to infer the spatial distributions of the ratios of OC to BC. Since these ratios determine the relative amounts of scattering and absorption, they are often used to estimate the radiative forcing due to aerosols. An artifact in the protocol for filter measurements of OC has led to widespread overestimates of the ratio of OC to BC in atmospheric aerosols. We developed a criterion to correct for this artifact and analyze corrected OC to BC ratios. The OC to BC ratios, ranging from 1.3 to 2.4, appear relatively constant and are generally unaffected by seasonality, sources, or technology changes, at the locations considered here. The ratios compare well with emission inventories over Europe and China but are a factor of 2 lower in other regions. The reduced estimate for OC/BC in aerosols strengthens the argument that reduction of soot emissions maybe a useful approach to slow global warming.

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1. Introduction

[2] Carbonaceous aerosols produced by incomplete combustion of fossil and biomass fuels are ubiquitous in the atmosphere at concentrations comparable to sulfates, the major inorganic aerosol species. Sulfate aerosols influence climate through their light-scattering properties. Carbonaceous aerosols are composed of both light-absorbing black carbon (BC) and light-scattering organic carbon (OC). In this paper, we are concerned with the relative amounts of OC and BC in aerosol particles, particularly as it relates to the radiative forcing of climate.

[3] Whether carbonaceous particles principally warm or cool the atmosphere depends on the aerosol single scattering albedo (SSA), which is the ratio of incident radiation that is scattered to the incident radiation that is absorbed. It follows that the aerosol SSA depends on the relative amounts of absorbing BC and scattering OC. An artifact in the filter measurements of OC has led to significant overestimates of the ratio of OC to BC in atmospheric aerosols. We developed a procedure to correct the published OC/BC ratios for this artifact. The corrected ratios are significantly lower than reported in the literature. This implies that carbonaceous particles may have lower OC/BC ratios, have lower SSA than previously assumed, and thus will more likely have a warming effect on climate.

[4] The net effect of the radiative forcing of climate by carbonaceous aerosols is assessed by modeling studies. In climate models, predicted OC/BC ratios are based on OC and BC emissions estimated from sector segregated fuel consumption and relevant OC and BC emission factors [*Cooke et al.*, 1999; *Bond et al.*, 2004]. Estimated emission inventories of both OC and BC, and the derived OC/BC ratios, are subject to significant uncertainties in both the fuel data and especially the adopted emission factors, signifying the need to compare the inventory-based and measured OC/BC ratios.

[5] Measured OC and BC concentrations values, and therefore the OC/BC ratios, are also subject to uncertainties. These uncertainties are the result of sampling and analytical artifacts, which tend to either overestimate or underestimate the concentrations of both OC and BC. OC concentrations are commonly determined by analysis of particles collected with quartz filters. During sample collection, gaseous organic compounds present in the sampled air adsorb onto the filter material. This process, known as the positive artifact, increases the OC of the filter deposit and thus the OC/BC ratio [*Kirchstetter et al.*, 2001, and references therein]. Thus, measured OC emissions, as well as the OC/BC ratio, will be artificially high if the positive OC sampling artifact is overlooked. For example, uncorrected measurements

Table 1. Data Sources Used in Analysis[a]

Location	Sampling Dates	Particle Size Cut, μm/ Analytical Method	OC, μg m^{-3}	BC, μg m^{-3}	OC/BC	References
China						
Beijing 1	Summer 1999	2.5/TO	13.42	6.27	2.14	*He et al.* [2001]
Beijing 2	Fall 1999	2.5/TO	28.79	10.23	2.81	*He et al.* [2001]
Beijing 3	Winter 1999–2000	2.5/TO	31.49	11.08	2.84	*He et al.* [2001]
Beijing 4	Spring 2000	2.5/TO	18.21	6.67	2.73	*He et al.* [2001]
Beijing 5	Annual 1999–2000	2.5/TO	25.30	9.40	2.69	*He et al.* [2001]
Shanghai 1	Hainan, summer 1999	2.5/TO	13.07	5.71	2.29	*Ye et al.* [2003]
Shanghai 2	Hainan, fall 1999	2.5/TO	17.28	6.97	2.48	*Ye et al.* [2003]
Shanghai 3	Hainan, winter 1999–2000	2.5/TO	17.59	8.07	2.18	*Ye et al.* [2003]
Shanghai 4	Hainan, annual 1999–2000	2.5/TO	15.98	6.92	2.31	*Ye et al.* [2003]
Shanghai 5	Tongji, spring 1999	2.5/TO	16.10	5.27	3.06	*Ye et al.* [2003]
Shanghai 6	Tongji, summer 1999	2.5/TO	9.62	4.61	2.09	*Ye et al.* [2003]
Shanghai 7	Tongji, fall 1999	2.5/TO	15.22	6.81	2.23	*Ye et al.* [2003]
Shanghai 8	Tongji, winter 1999–2000	2.5/TO	16.40	8.16	2.01	*Ye et al.* [2003]
Shanghai 9	Tongji, annual 1999–2000	2.5/TO	14.34	6.21	2.31	*Ye et al.* [2003]
Hong Kong 1	PU site, Nov–Feb 2000–01	2.5/MNO	9.45	5.80	1.63	*Ho et al.* [2003]
Hong Kong 2	KT site, Nov–Feb 2000–01	2.5/MNO	10.16	5.05	2.01	*Ho et al.* [2003]
Hong Kong 3	HT site Nov–Feb 2000–01	2.5/MNO	5.52	1.36	4.06	*Ho et al.* [2003]
Hong Kong 4	Average 3 sites	2.5/MNO	8.38	4.07	2.06	*Ho et al.* [2003]
Hong Kong 5	Jan–Feb 2002	2.5/TO	9.60	4.70	2.04	*Cao et al.* [2003]
Shenzen	Jan–Feb 2002	2.5/TO	13.20	6.10	2.16	*Cao et al.* [2003]
Guangzhou,	Jan–Feb 2002	2.5/TO	22.60	8.30	2.72	*Cao et al.* [2003]
Zhuhai	Jan–Feb 2002	2.5/TO	12.20	5.00	2.44	*Cao et al.* [2003]
Pearl river delta	All sites Jan–Feb 2002	2.5/TO	14.70	6.10	2.41	*Cao et al.* [2003]
Macao	Dec Jul 2001, Dec 2002	2.5/TO	12.20	4.40	2.77	*Wu et al.* [2003]
Hong Kong 6	9 site average 1998–2001	10/TO	8.89	4.66	1.91	*Yu et al.* [2004]
Beijing 6	Jun–Jul 2002	2.5/2ST	10.70	5.70	1.88	*Dan et al.* [2004]
Beijing 7	Dec 2002	2.5/2ST	36.70	15.20	2.41	*Dan et al.* [2004]
Beijing 8	Summer 2002	2.5/2ST	12.40	5.40	2.29	*He et al.* [2004]
Relative standard deviation (%)			47	40	20	
Japan						
Sapporo 1	Annual 1982	8.0/2ST	4.15	5.10	0.81	*Ohta and Okita* [1984]
Chichi-Jima	Dec 1981	TSP/2ST	0.60	0.70	0.86	*Ohta and Okita* [1984]
Hachijo-Jima 1	Jan 1981	TSP/2ST	1.40	1.00	1.40	*Ohta and Okita* [1984]
Hachijo-Jiima 2	Jan 1981	TSP/2ST	1.00	0.70	1.43	*Ohta and Okita* [1984]
Nagoya	Average 1984–1986	TSP/2ST	16.20	13.00	1.25	*Kadowaki* [1990]
Tsushima	1991	na	2.32	1.55	1.50	*Hatakeyama* [1993]
Oki Island	1991	na	1.60	1.25	1.28	*Hatakeyama* [1993]
Okinawa	1991	na	0.78	1.17	0.67	*Hatakeyama* [1993]
Nagano	1991	na	1.20	1.50	0.80	*Hatakeyama* [1993]
Sapporo 2	Site 1, Jun 1987–Dec 1988	10/2ST	3.40	3.30	1.03	*Kaneyasu et al.* [1995]
Sapporo 3	Site 2, Jun 1987–Dec 1988	10/2ST	3.60	3.60	1.00	*Kaneyasu et al.* [1995]
Sapporo 4	Annual 1991	10/2ST	3.74	4.26	0.88	*Ohta et al.* [1998]
Uji	Jan 1989–Nov 1999	10/RP	2.03	2.64	0.77	*Holler et al.* [2002]
Tokyo	Dec 1998–Jan 1999	2.5/RP	7.80	5.40	1.44	*Saitoh et al.* [2002]
Kyoto	Summer 2002	2.5/2ST	5.00	1.30	3.85	*He et al.* [2004]
Relative standard deviation (%)			109	102	61	
K Korea and other Asian countries						
Kosan 1	Jul 1994	2.5/MNO	4.58	0.38	12.05	*Kim et al.* [1998]
Kosan 2	Aug 1994	2.5/MNO	2.36	0.08	29.50	*Kim et al.* [1998]
Seoul 1	Jun 1994	2.5/MNO	9.97	7.57	1.32	*Kim et al.* [1999]
Chongju	Annual 1995–1996	2.5/TO	4.99	4.44	1.12	*Lee and Kang* [2001]
Sihwa	1998–1999	2.5/MNO	9.1	1.8	5.06	*Park et al.* [2001]
Kosan 3 (Cheju isl)	Mar 96	2.5/MNO	2.97	0.32	9.28	*Lee et al.* [2001]
Kosan 4 (Cheju isl)	Dec 1996	2.5/MNO	4.41	0.43	10.26	*Lee et al.* [2001]
Kosan 5 (Cheju isl)	Jan 1997	2.5/MNO	3.31	0.23	14.39	*Lee et al.* [2001]
Kosan 6 (Cheju isl)	Sep 1997	2.5/MNO	3.56	0.42	8.48	*Lee et al.* [2001]
Kosan 7(Cheju isl)	Dec 1997	2.5/MNO	2.6	0.34	7.65	*Lee et al.* [2001]
Kosan 8 Average	Annual average 1996–1997	2.5/MNO	3.26	0.34	9.59	*Lee et al.* [2001]
Kanghwa 1	Mar 1996	2.5/MNO	5.16	0.56	9.21	*Lee et al.* [2001]
Kanghwa 2	Dec 1996	2.5/MNO	11	2.45	4.49	*Lee et al.* [2001]
Kanghwa 3	Jan 1997	2.5/MNO	7.58	0.95	7.98	*Lee et al.* [2001]
Kanghwa 4	Sep 1997	2.5/MNO	4.28	0.79	5.42	*Lee et al.* [2001]
Kanghwa 5	Dec 1997	2.5/MNO	6.23	0.9	6.92	*Lee et al.* [2001]
Kanghwa 6	Annual average 1996–1997	2.5/MNO	6.45	0.98	6.58	*Lee et al.* [2001]
Seoul 2	Nov–Dec 1999	2.5/MNO	15.2	7	2.17	*Park et al.* [2002]
Kwangju	Jun 2000	2.5/MNO	7.6	5.3	1.43	*Park et al.* [2002]
Gwangju	Summer 2002	2.5/2ST	1.4	0.3	4.67	*He et al.* [2004]
Lahore	1992–1993	TSP/TO	74.7	17.53	4.26	*Smith et al.* [1996]
Mumbai	Mar 1999	10/TO	25.3	12.6	2.01	*Venkataraman et al.* [2002]

Table 1. (continued)

Location	Sampling Dates	Particle Size Cut, μm/ Analytical Method	OC, μg m^{-3}	BC, μg m^{-3}	OC/BC	References
Dhaka	Apr–May 2001	TSP/2ST	45.7	22	2.08	*Salam et al.* [2003a]
Bhola 1	May 14–17, 2001	TSP/2ST	4.22	3.18	1.33	*Salam et al.* [2003b]
Bhola 2	May 1–21, 2001	TSP/2ST	5.04	2.37	2.13	*Salam et al.* [2003b]
Ulan Bator	Jul 2002	2.5/2ST	2.3	0.4	5.75	*He et al.* [2004]
Relative standard deviation (%)			124	156	89	
Europe						
Athens 1	Jun–Aug 1982	TSP/TO	25.7	8.2	3.13	*Valaoras et al.* [1988]
Athens 2	Jan–Feb 1983	TSP/TO	16.1	11	1.46	*Valaoras et al.* [1988]
Paris 1	Fall 1984	TSP/2ST	4.2	1.6	2.63	*Cachier et al.* [1989]
Paris 2	Winter 1985	TSP/2ST	14.6	5.9	2.47	*Cachier et al.* [1989]
Gif sur Yvette 1	Winter 1986	TSP/2ST	6.7	2.4	2.79	*Cachier et al.* [1989]
Gif sur Yvette 2	Spring 1986	TSP/2ST	6.5	1.9	3.42	*Cachier et al.* [1989]
Gif sur Yvette 3	Summer 1986	TSP/2ST	4.8	1.5	3.20	*Cachier et al.* [1989]
Gif sur Yvette 4	Fall 1985	TSP/2ST	18.9	4	4.73	*Cachier et al.* [1989]
Areao	Nov 1993–Aug 1994	0.95/TO	2.73	0.82	3.33	*Pio et al.* [1996]
Birmingham,	May 1993	TSP/TO	4.82	1.38	3.49	*Castro et al.* [1999]
Tábua	Jul 1994–Aug 1995	TSP/TO	5.83	1.17	4.98	*Castro et al.* [1999]
Anadia	Aug 1996	TSP/TO	3.5	1.6	2.19	*Castro et al.* [1999]
Birmingham,	Jan 1994	TSP/TO	4.78	3.42	1.40	*Castro et al.* [1999]
London 1	Site 1, 1995	TSP/TO	7.6	2.6	2.92	*Kendall et al.* [2001]
London 2	Site 2, 1995	TSP/TO	6.3	2	3.15	*Kendall et al.* [2001]
Aspvreten	Jun 1996	10/2ST	2.2	0.1	22.00	*Zappoli et al.* [1999]
S. Pietro Capofiume	Sep 1996	10/2ST	6.2	1	6.20	*Zappoli et al.* [1999]
K-Puszta 1	Jul 1996	10/2ST	5	0.6	8.33	*Zappoli et al.* [1999]
K-Puszta 2	Jul–Aug 1996	2.5/RP	10.6	0.42	25.24	*Molnár et al.* [1999]
Coimbra 1	Oct 1992–Mar 1993	TSP/TO	8.88	4.22	2.10	*Castro et al.* [1999]
Coimbra 2	Aug–Sep 1993	TSP/TO	5.32	1.78	2.99	*Castro et al.* [1999]
Oporto 1	Apr–Sep 1993	TSP/TO	7.22	2.67	2.70	*Castro et al.* [1999]
Oporto 2	Oct 1992–Mar 1993	TSP/TO	9.06	5.33	1.70	*Castro et al.* [1999]
Aveiro	Aug 1996	TSP/TO	3.04	1.26	2.41	*Castro et al.* [1999]
Basel 1	Winter 1997–1998	10/2ST O	5.15	3.47	1.48	*Röösli et al.* [2001]
Basel 2	Spring 1997–1998	10/2ST O	3.5	2.2	1.59	*Röösli et al.* [2001]
Basel 3	Summer 1997–1998	10/2ST O	4.43	2.86	1.55	*Röösli et al.* [2001]
Basel 4	Fall 1997–1998	10/2ST O	5.44	4.55	1.20	*Röösli et al.* [2001]
S. Pietro Capofiume	Jan 1998–Apr 1999	1.5/T	8.61	0.53	16.25	*Decesari et al.* [2001]
Helsinki	2000–2001	2.5/TO	2.96	1.15	2.73	*Viidanojaa et al.* [2002]
Melpitz	Apr 29–May 5, 2001	10/TO	7.7	2.60	2.96	*Carvalho et al.* [2003]
Budapest	23 Apr–5 May, 2002	2.5/TO	6.8	3.40	2.00	*Salma et al.* [2004]
Relative standard deviation (%)			69	85	123	
Relative standard deviation (%) for BC > 1.5 μg m^{-3}			67	63	36	
North America						
Denver, CO	Nov/Dec 1978	2.0/T	7.5	6.4	1.17	*Countess et al.* [1981]
New York, NY	1978/79	3.5/TO	5.14	3.12	1.65	*Shah et al.* [1985]
Detroit, MI 1	Jul 1981	2.5/T	7.1	1.6	4.44	*Wolff and Korsog* [1985]
Detroit, MI 2	Jun–Jul 1981	2.5/T	5.38	2.32	2.32	*Wolff et al.* [1985]
Allegheny Mtn.,	Aug 1983	5.5/TO	2	1.2	1.67	*Keeler et al.* [1986]
Laurel Hill, MD	Aug 1983	5.5/TO	2.3	1.4	1.64	*Keeler et al.* [1986]
Lewes, DE 1	Feb 1983	2.5/T	2.4	1.1	2.18	*Wolff et al.* [1986]
Phoenix, AZ	Jan 1983	2.8/2ST	10	8.3	1.20	*Solomon and Moyers* [1986]
No. Michigan 1	Dec 1984–Apr 1985	TSP/T	2.2	0.48	4.58	*Cadle and Dasch* [1988]
No. Michigan 2	Dec 1983–Apr 1984	TSP/T	1.72	0.72	2.39	*Cadle and Dasch* [1988]
Southern Ontario	Jul 1986	2.5/TO	5.2	0.8	6.50	*Keeler et al.* [1990]
Lewes, DE 2	Aug 1982	2.5/T	4	0.7	5.71	*Wolff et al.* [1986].
Angeles Natl. Forest, CA	Annual, 1986	10/TO	6.4	1.2	5.33	*Solomon et al.* [1989]
Detroit, MI 3	Jan–Apr 1985	TSP/2ST	5.7	2.1	2.71	*Muhlbaier-Dasch and Cadle* [1989]
Detroit, MI 4	Jan–Mar 1984	TSP/2ST	5.5	2.4	2.29	*Muhlbaier-Dasch and Cadle* [1989]
Crows Landing, SJV, CA	Annual 1988–1089	2.5/TO	3.3	1.46	2.26	*Chow et al.* [1993a, 1993b]
Kern, SJV, CA	Annual 1988–1089	2.5/TO	2.66	1.32	2.02	*Chow et al.* [1993a, 1993b]
Stockton, CA	Annual 1988–1089	2.5/TO	5.42	3.85	1.41	*Chow et al.* [1993a, 1993b]
Bakersfield, CA	Annual 1988–1089	2.5/TO	6.5	5.44	1.19	*Chow et al.* [1993a, 1993b]
Fresno, CA	Annual 1988–1089	2.5/TO	8.05	6.27	1.28	*Chow et al.* [1993a, 1993b]
Galveston, background	Annual 1997–1998	2.5/TO	1.8	0.7	2.57	*Fraser et al.* [2002]
HRM, supersite, TX	Annual 1997–1998	2.5/TO	3.3	1.7	1.94	*Fraser et al.* [2002]
Bingle, suburban, TX	Annual 1997–1998	2.5/TO	4	2	2.00	*Fraser et al.* [2002]
Clinton, suburban, TX	Annual 1997–1998	2.5/TO	3.7	2.1	1.76	*Fraser et al.* [2002]
Fort Meade, MD	Jul 1999–Mar 2000	2.5/TO	1.43	1.06	1.35	*Chen et al.* [2002]
Mexico City 1	Site 1, Feb–Mar 1997	2.5/TO	9.82	4.65	2.11	*Chow et al.* [2002]
Mexico City 2	Site 2, Feb–Mar 1997	2.5/TO	11	5.59	1.97	*Chow et al.* [2002]
Mexico City 3	Site 3, Feb–Mar 1997	2.5/TO	12.02	9.39	1.28	*Chow et al.* [2002]
Mexico City 4	Site 4, Feb–Mar 1997	2.5/TO	9.98	8.28	1.21	*Chow et al.* [2002]

Table 1. (continued)

Location	Sampling Dates	Particle Size Cut, μm/ Analytical Method	OC, $\mu g\ m^{-3}$	BC, $\mu g\ m^{-3}$	OC/BC	References
Mexico City 5	Site 5, Feb–Mar 1997	2.5/TO	7.62	2.89	2.64	*Chow et al.* [2002]
Mexico City 6	Site 6, Feb–Mar 1997	2.5/TO	8.53	3.75	2.27	*Chow et al.* [2002]
Look Rock TN 1	Fall 2001	2.5/TO	3.36	0.55	6.11	*Tanner et al.* [2004]
Lawrence co, TN 1	Winter 2001	2.5/TO	2.43	0.64	3.80	*Tanner et al.* [2004]
Look Rock 2	Winter 2001	2.5/TO	1.86	0.65	2.86	*Tanner et al.* [2004]
Look Rock 3	Spring 2001	2.5/TO	3.57	0.66	5.41	*Tanner et al.* [2004]
Look Rock 4	Summer 2001	2.5/TO	4	0.66	6.06	*Tanner et al.* [2004]
Lawrence co 2	Summer 2001	2.5/TO	4.28	0.74	5.78	*Tanner et al.* [2004]
Lawrence co 3	Spring 2001	2.5/TO	3.57	0.83	4.30	*Tanner et al.* [2004]
Lawrence co 4	Fall 2001	2.5/TO	4.42	0.91	4.86	*Tanner et al.* [2004]
Chattanooga 1	Summer 2001	2.5/TO	3.71	1.1	3.37	*Tanner et al.* [2004]
Chattanooga 2	Spring 2001	2.5/TO	5.71	1.6	3.57	*Tanner et al.* [2004]
Chattanooga 3	Fall 2001	2.5/TO	7.35	2.2	3.34	*Tanner et al.* [2004]
Chattanooga 4	Winter 2001	2.5/TO	5.07	2.4	2.11	*Tanner et al.* [2004]
Relative standard deviation (%)			53	94	55	
Relative standard deviation (%) for BC > 1.5 $\mu g\ m^{-3}$			34	60	41	

[a]Designations and description of analytical methods given in text.

during ACE-Asia project overestimated particle-phase OC by 20 to 100% [*Mader et al.*, 2003]. Measured OC/BC ratios in biomass smoke plumes in Southern Africa were nearly twice as large if OC was not corrected for the positive artifact [*Kirchstetter et al.*, 2003].

[6] Additionally, some of the volatile components of the particle phase OC may be desorbed from the filter during sampling. This process is known as the negative OC sampling artifact and results in an underestimation of OC concentrations [*Eatough et al.*, 1999].

[7] The main uncertainty in introduced by the analytical methods pertains to their ability to accurately separate BC and OC [*Schmid et al.*, 2001] and, therefore, bias the published OC and BC values. The repercussions of these analytical problems on our findings are discussed in the subsequent sections of this paper.

[8] Another uncertainty relates to the fact that commonly used analytical methods quantify aerosol OC as the mass of carbon (generally expressed in units of $\mu g\ C\ m^{-3}$) content of the organic aerosol material (OM). The mass of (OM) is larger than OC because OM contains other species besides carbon. As the OM is the relevant quantity for climate studies its concentration is often approximated by OC concentrations multiplied by a constant factor. Climate models, for example, commonly assume that OM is 1.3 or 1.4 times greater than the OC. The value of the factor relating OM and OC, however, is not accurately known [*Turpin and Lim*, 2001]. In the following, we consider OC, since it is the quantity that is actually measured.

[9] Here we present an analysis of OC/BC ratios derived from published OC and BC concentrations measured in mostly urban and some non-urban locations in Asia, Europe and North America. Most of these data have not been corrected for the positive OC sampling artifact. Therefore, in the following discussion we refer to these as "apparent" concentrations and ratios. Accordingly, after presenting the published data, we describe a procedure to distinguish between data that is positive artifact-affected and positive artifact unaffected based on the nature of the OC sampling artifact. We then use unaffected data to infer regional and

seasonal OC/BC ratios and make comparisons with values of OC/BC based on emission inventories.

2. Results

[10] The OC and BC concentration data used in this study were obtained from filter samples analyzed by a number of investigators using different analytical methods. These data are taken from the literature and arranged roughly by year of publication (Table 1). They include long term (i.e., annual) and short term (i.e., daily and weekly) averaged concentrations measured at sites affected by different fuel types, fuel consumption, and combustion technologies. Table 1 also indicates the analytical methods and aerosol size cuts employed in sample collection. Analytical methods denoted as "TO" combine thermal an optical measurement approach to improve OC – BC separation. "T" stands for solely thermal, usually temperature programmed analysis. Two-step methods "2ST" relay on exposing the sample to a fixed temperature, chosen to remove the OC from the sample. Carbon content of the exposed sample is operationally defined as BC, and the difference between the total carbon of unexposed and exposed sample gives the OC. "MNO" method uses MnO_2 as the oxygen donor for carbon oxidation at several pre-set temperatures. The details of the methods are in the references listed in Table 1.

[11] To compare objectively the OC/BC ratios and BC concentrations, the relative standard deviations (rsd = 100% * standard deviation/average) of OC, BC, and OC/BC are included in Table 1 for each region considered.

2.1. Spatial and Seasonal OC/BC Variations

[12] OC/BC ratios for sites in China (Figure 1a) are nearly constant (average 2.39 ± 0.47) irrespective of large variations in the range of BC concentration (4 μg BC m^{-3} to 15 μg BC m^{-3}). OC/BC ratios appear to be the same in Beijing and Shanghai (2.39 ± 0.32), Pearl River Delta sites (2.44 ± 0.28), and Hong Kong (2.28 ± 0.88).

[13] OC/BC ratios in Japan are similar in urban, coastal and island sites (Figure 1b). This insensitivity to location
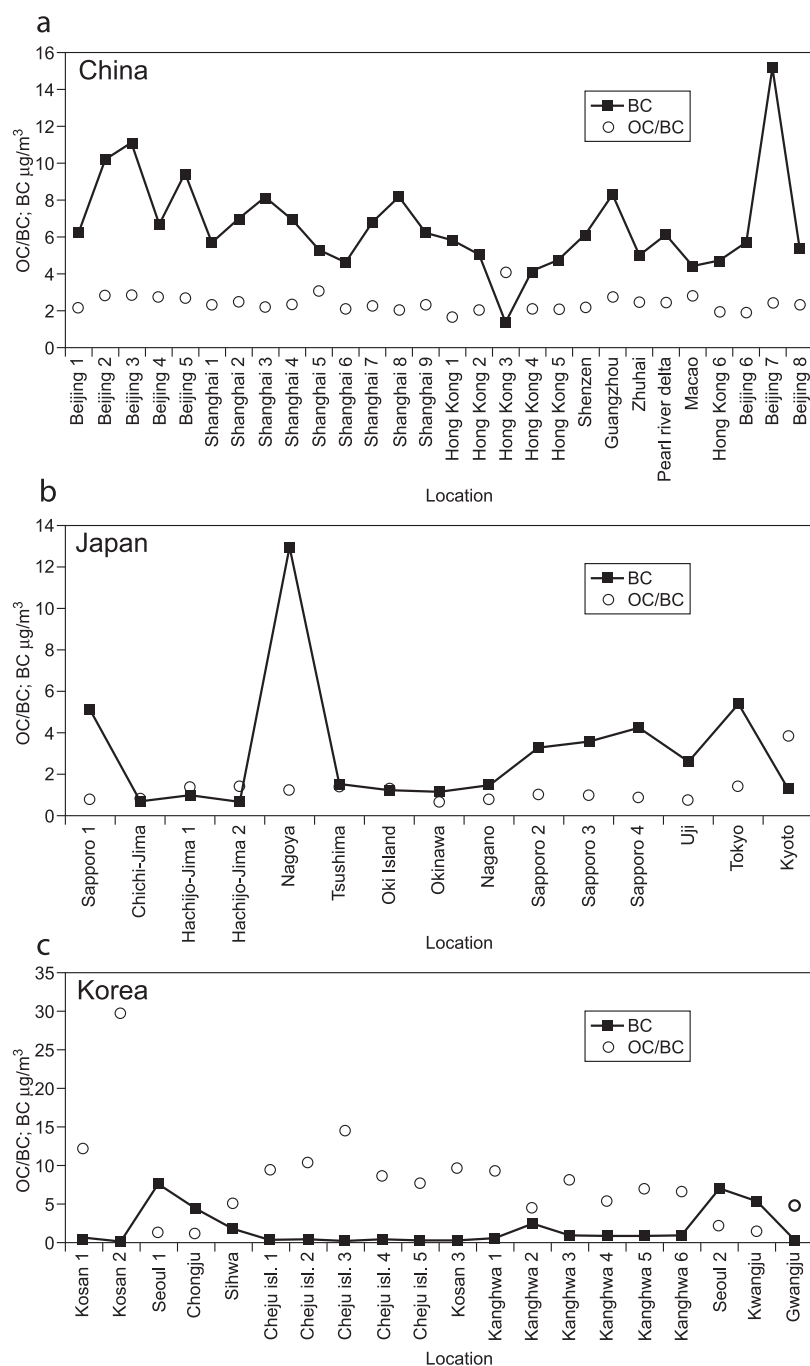
Figure 1. Variations of BC concentrations and apparent OC/BC ratios for (a) China, (b) Japan, and (c) Korea and other Asia.

and BC concentrations is qualitatively similar to that observed in China (Figure 1a). However, the average OC/BC value for Japan (1.10 ± 0.31) is lower than measured in any other country we studied. The low value of this ratio in Japan suggests that BC and OC emissions may be primarily derived from automotive sources, for which the OC/BC ratios are known to be low. For example, OC/BC ratios of automotive emissions in California from diesel and gasoline engines are 0.5 ± 0.4 and 0.9 ± 0.4, respectively [*Kirchstetter et al.*, 2004]. The OC/BC ratio measured in a tunnel in Austria is similar [*Laschober et al.*, 2004].

[14] OC/BC ratios for Korea and other Asian sites are distinctly different at continental and coastal sites (Figure 1c). For Korean mainland sites the average value of OC/BC (1.51 ± 0.46) falls between those for China and Japan. The OC/BC ratios for Korean island and coastal locations, however, are considerably higher. Average OC/BC ratio for Pakistan, and Mongolia (≈5) is substantially higher than the average for several cities in India and Bangladesh (≈1.9).

[15] Plots of OC/BC distributions for European and North American locations are shown in Figure 2. European data
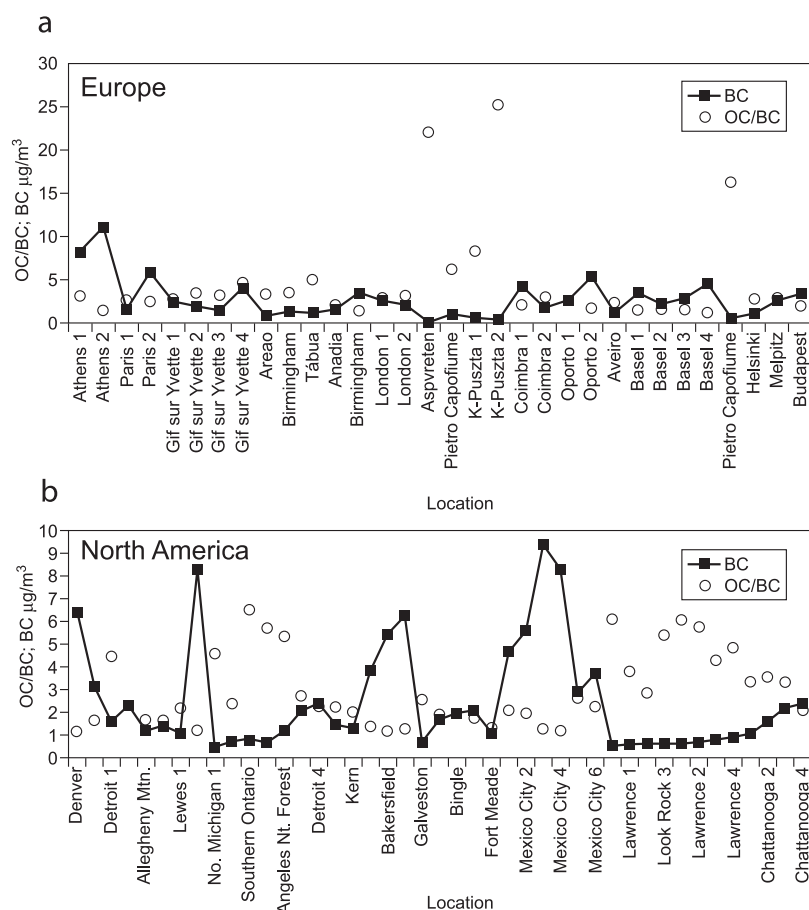
Figure 2. Same as in Figure 1, but for (a) Europe and (b) North America.

(Figure 2a) have an average OC/BC of 2.69 ± 0.91 for urban locations, increasing to values as high as 25 for regional background locations in Hungary, Italy, and Sweden, where BC concentrations are low. The average OC/BC ratio (2.94 ± 1.61) for North American locations (Figure 2b) is similar to the European ratios.

[16] Finally, we note that BC concentrations at urban locations, such as Beijing, Shanghai, Chongju, Sapporo, and Basel, show a significant increase in winter. However, no corresponding seasonal change in OC/BC ratios is observed at these locations (Table 2). At a few other locations such as Cheju Island and Gif sur Yvette (Table 1) there might be indications of seasonal differences in the OC/

BC ratios. At these sites, however, seasonal BC variations are less pronounced, and BC concentrations are much lower than in the examples shown in Table 2.

2.2. OC/BC Ratios and BC Concentrations

[17] As indicated above, the apparent OC/BC ratios for China and Japan (Figures 1a and 1b) do not depend appreciably on BC concentration. However, the OC/BC ratios for Korea (Figure 1c), Europe, and North America, (Figures 2a and 2b) are highly variable and appear to depend on BC concentration. High ratios are systematically associated with the lowest BC concentrations, and low ratios with the highest BC concentrations.

Table 2. Seasonal Changes in OC/BC Ratios and BC Concentrations ($\mu g\ m^{-3}$)

|	Winter	Spring	Summer	Fall	Average
Location	OC/BC (BC)	OC/BC (BC)	OC/BC (BC)	OC/BC (BC)	OC/BC
Beijing[a] 1999–00	2.84 (11.08)	2.73 (6.67)	2.14 (6.27)	2.81 (10.23)	2.63 ± 0.33
Shanghai[b] 1999	2.01 (8.16)	3.05 (5.27)	2.09 (4.61)	2.23 (6.81)	2.34 ± 0.46
Chongju[c] 1995–96	1.16 (4.32)	1.34 (3.59)	1.20 (3.37)	0.94 (6.35)	1.16 ± 0.16
Sapporo[d] 1998	0.64 (7.0)	0.82 (3.97)	0.83 (3.03)	1.06 (6.13)	0.84 ± 0.17
Basel[e] 1997–98	1.48 (3.47)	1.59 (2.2)	1.55 (2.86)	1.20 (4.55)	1.41 ± 0.18

[a]*He et al.* [2001].
[b]*Ye et al.* [2003].
[c]*Lee and Kang* [2001].
[d]*Ohta and Okita* [1984].
[e]*Röösli et al.* [2001].
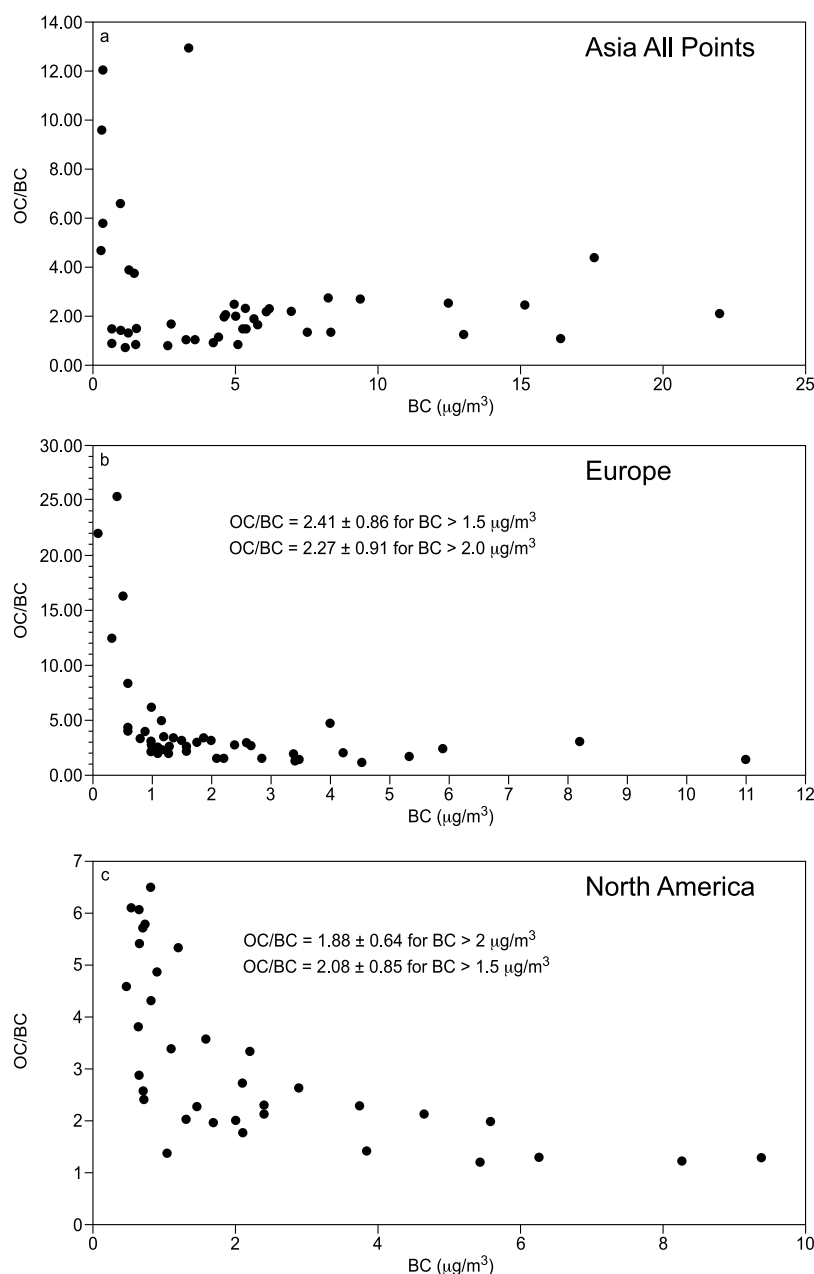
Figure 3. Plots of apparent OC/BC ratios versus BC concentrations for (a) Asia, (b) Europe, and (c) North America.

[18] The OC/BC versus BC plots for Asia, Europe, North America, and other locations illustrate this trend. A plot of all Asian data (Figure 3a) shows that high ratios (mostly for coastal Korea) are clustered at BC concentrations <1 μg m^{-3}. Low ratios for Japan (≈ 1.0), urban Korea (≈ 1.5), all of China (≈ 2) and other Asian locations correspond to high BC concentrations. European data (Figure 3b) show similar features: extremely high ratios (up to ≈ 25) for BC < 1 μg m^{-3} at "background" locations in Italy, Hungary, and Sweden, and ratios of 2.41 ± 0.86 for BC concentrations >1.5 μg m^{-3}. In North America (Figure 3c) half of all data points that show considerable scatter and relatively large ratios are for BC < 2 μg m^{-3}. The average OC/BC ratio for BC concentrations above 2 μg m^{-3} is

1.88 ± 0.64. Average OC/BC for all data considered here (Table 1) is 3.32 ± 3.48, whereas this ratio drops to 2.20 ± 1.51 for BC concentrations >1.5 μg m^{-3}.

3. Discussion
3.1. Correction Criteria

[19] The results presented above demonstrate that high apparent OC/BC values systematically occur only with low BC concentrations. Next we show that neglect of the positive sampling artifact, which causes an overestimation of OC, is responsible for the observed OC/BC versus BC relationship.
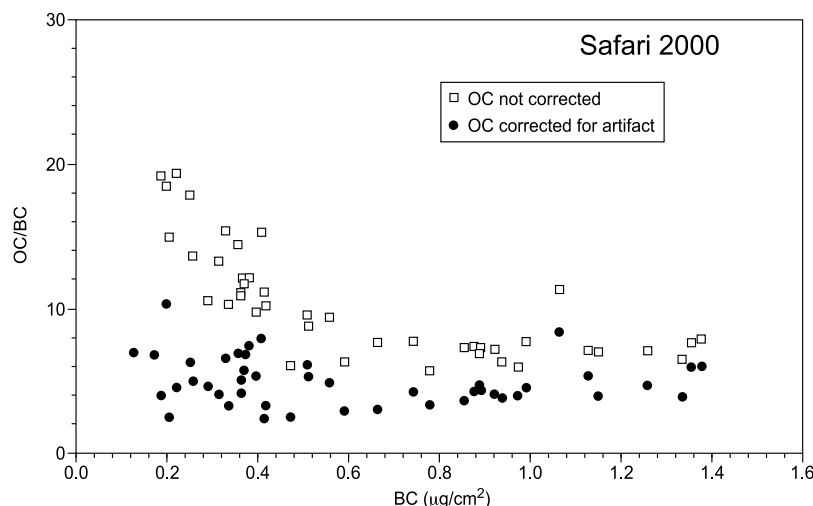
Figure 4. Comparison of uncorrected and positive artifact corrected OC/BC ratios measured during SAFARI 2000.

[20] The dependence of the apparent OC/BC ratios on BC concentrations is expected from the positive artifact mechanism. The magnitude of this artifact is related to the limited capacity of the filter for retaining the adsorbed (artifact) gasphase OC. As a consequence, the loading of adsorbed OC becomes a smaller fraction of total OC when the amount of gaseous (and particulate) species passing through the filter is high [*Kirchstetter et al.*, 2001; Lunden et al., manuscript in preparation].

[21] Positive artifact corrected OC concentration can be obtained by sampling with two quartz filters placed back-to-back as described by *Kirchstetter et al.* [2001] and *Mader et al.* [2003]. The top, or front, filter removes all particles, while gas-phase organics adsorb onto both the front and back filters. Thus, the measured carbon content of the backup filter can be subtracted from that of the front filter to give an estimate of the particle phase OC. (Uncorrected or apparent OC/BC is derived from the front filter only.)

[22] That the observed high OC/BC ratios at low BC concentrations are greatly overestimated is demonstrated by comparing the uncorrected ("apparent") and positive artifact-corrected OC/BC ratios. During the SAFARI 2000 project [*Swap et al.*, 2003], OC and BC concentrations were determined for a large number of aircraft collected samples taken in and out of biomass burning plumes [*Kirchstetter et al.*, 2003]. The OC concentrations reported in that study were corrected for artifact OC by the method mentioned above. Figure 4 shows the different dependence of artifact-corrected and uncorrected ratios on BC concentration. The uncorrected SAFARI ratios are as high as 27 at the lowest BC concentrations, and asymptotically approach the corrected value of about 6 at BC concentrations >1 μg cm^{-2}.

[23] Artifacts inherent in some analytical methods may, in addition to positive and negative sampling artifacts, cause erroneous OC/BC ratios. Thermal analysis methods rely on heating a sample in an oxidizing or inert atmosphere and measuring the gaseous species evolved from the sample as a function of the sample temperature. This enables a separa-

tion of the carbonaceous material according to their volatilization, decomposition and combustion characteristics. The component that evolves at the highest temperature in then operationally defined as the black or elemental carbon. However, if a char-like material has been formed by OC pyrolysis then this fraction may be erroneously analyzed as the BC and thus cause a lower OC/BC value. One way to correct for the charring artifact is to perform optical transmission or reflectance measurement on the sample simultaneously with thermal analysis. Monitoring the optical signal allows for following the charring process and to more accurately quantify the actual BC and OC content and the OC/BC ratio.

[24] We can evaluate the effect of the charring artifact on OC/BC ratios by considering a set of data (from Table 1) for Texas, Tennessee, and Mexico City [*Fraser et al.*, 2002; *Tanner et al.*, 2004; *Chow et al.*, 2002]. All of these data were obtained by the same analytical method Thermal Optical Reflection (TOR) [*Chow et al.*, 1993a, 1993b], that corrects for the charring effect. An examination of this data set shows that the for low BC concentrations (<2 μg m^{-3}) the average OC/BC is 4.22 ± 1.42. At higher BC concentrations (>2 μg m^{-3}), however, the average OC/BC is 1.90 ± 0.64. This trend is consistent with the trend of apparent OC/BC ratios, such as shown in Figure 3, suggesting that accounting for the charring artifact does not alter our positive sampling artifact based interpretation of the OC/BC versus BC trend.

[25] Lastly, we note that the data considered are insufficient to evaluate the effect of negative sampling artifact on OC/BC ratios.

3.2. Corrected OC/BC Ratios

[26] Based on the foregoing discussion we can identify apparent OC/BC ratios that can be used as "substitutes" for the artifact-corrected values. These unaffected ratios do not significantly depend on BC concentration variations. For example, all ratios for China and Japan (Figures 1a and 1b) satisfy this criterion. For the European (Figure 2a),

Table 3. Comparison of Average BC Concentrations and Apparent and Corrected OC/BC Ratios

Region	Average BC	Apparent OC/BC	Corrected OC/BC
China	7.10 ± 4.11	2.35 ± 0.61	2.35 ± 0.61
Japan	3.09 ± 5.17	1.29 ± 0.75	1.29 ± 0.75
Korea	3.85 ± 3.39	4.47 ± 4.9	1.47 ± 0.47[x]
Europe	2.22 ± 2.09	4.31 ± 5.0	2.41 ± 0.86
			2.27 ± 0.91
North America	2.46 ± 2.32	2.92 ± 1.59	2.08 ± 0.85
			1.88 ± 0.64
All sites	3.36 ± 3.69	3.32 ± 3.48	2.20 ± 1.51
			2.11 ± 1.56
INDOEX	2.71 ± 1.43	3.4 ± 1.7	1.40 ± 0.70
SAFARI		10.50 ± 3.41	6.06 ± 1.85

North American (Figure 2b), and composite Asian data (Figure 1c), we assume that the approximately constant OC/BC ratios, for BC concentrations >1.5 μg m^{-3} − 2.0 μg m^{-3}, represent the range of "correct" ratios.

[27] The apparent and corrected OC/BC ratios together with corresponding average BC concentrations (in μg m^{-3}) are summarized in Table 3. These data show that the unaffected (uncorrected) ratios can be lower than the apparent ratios by as much as a factor of two.

[28] A more quantitative evaluation of the relative constancy of OC/BC compared to BC, is provided by considering the relative standard deviations (rsd) of OC, BC, and OC/BC shown in Table 1. The rsd of the OC/BC ratio is about half of the rsd of BC (and OC) concentration for China, Japan, and Korea (and other Asian sites), supporting the conclusions given above. By contrast, the rsd of the OC/BC ratio is substantially larger than that of BC (and OC) for Europe, showing that OC/BC ratios are more variable than BC (and OC) in Europe. However, as asserted above, European OC/BC data are significantly overestimated when BC < 1.5 μg m^{-3}. Limiting the analysis to data from Europe where BC > 1.5 ugm^{-3} reveals that the RSD of the OC/BC ratio is about half that of BC, as observed for China, Japan, and Korea.

3.3. Comparison With Results From Emission Factors

[29] It is of interest to compare our derived ratios with OC and BC emission ratios calculated from emission factors and fuel consumption. The fuel and technology data used in such calculations are for entire countries, not for particular urban and non-urban locations where the measurements listed in this paper were made. We note, however, that the derived OC/BC ratios examined in this study do not show significant site-to-site and seasonal variations within a country, suggesting that average OC/BC approximately reflect the countrywide emissions. Furthermore, as the majority of these data (Table 1) were obtained in source dominated (urban) sites we may assume that the OC and BC at these sites are primary emission species.

[30] Table 4 compares calculated and measured (corrected) regional OC/BC ratios. Calculated ratios are based on OC and BC emissions (for 1996) separated into contained, fossil and biofuel, and open biomass burning [Bond et al., 2004]. As the data in Table 4 show, the measured and emission inventory based total OC/BC ratios agree for Europe and China. Measured values, however, are significantly lower for North America and India. Total

emission inventory based ratio for "Other Asian Countries" is more than twice the measured values for the three Asian countries and for INDOEX. These ratios are similar to the fossil fuel ratios, consistent with previous estimates of relative contributions of fossil and biomass sources for the Indian Ocean region [Mayol-Bracero et al., 2002; Novakov et al., 2000].

3.4. OC/BC Ratios and Radiative Forcing

[31] The relevance of artifact-corrected OC/BC ratios to the radiative forcing of climate by aerosols can be obtained by comparing them to those used in climate models. The carbonaceous aerosols in the models are commonly obtained either via a specification of the OC/BC ratio (e.g., Koch [2001] assumed OC/BC was 4 for fossil fuels and 8 for biomass sources) or via separate specification of OC and BC emission distributions. In either case, the resulting global distributions of carbonaceous aerosols will tend to overestimate the proportion of OC if the input data is not corrected for the positive sampling artifact. We compare in Figure 5, uncorrected values of observed OC/BC with the OC/BC ratios obtained in a current model, the Goddard Institute for Space Studies (GISS) ModelE [Schmidt et al., 2005]. The spatial distributions of the OC and BC emissions are based on Bond et al. [2004] for fossil fuel and bio fuel sources and on Van der Werf et al. [2004] for biomass sources. The OC/BC ratios derived from this model (in a simulation that includes both the direct and indirect aerosol effects from Menon and Del Genio [2005]) are compared to observed values in Figure 5a. In general, the model over estimates the ratios, with higher ratios predicted over most parts of the United States, India, parts of Asia, and over the

Table 4. Calculated and Corrected Measured Regional OC/BC Ratios

Region	Calculated[a]	Measured
North America		
Contained	1.50	
Biomass	12.69	
Total	4.11	1.88−2.08
United States		
Contained	1.52	
Biomass	14	
Total	3.4	1.88−2.08
Europe		
Contained	1.22	
Biomass	11.71	
Total	2.40	2.27−2.41
China		
Contained	1.54	
Biomass	5.75	
Total	1.89	2.35
India		
Contained	2.82	
Biomass	5.49	
Total	3.25	2.01
Other Asia		
Contained	1.84	
Biomass	7.13	
Total	3.29	Japan 1.29
		Korea 1.47
		Bangladesh 1.84
		INDOEX 1.40

[a]From Bond et al. [2004].
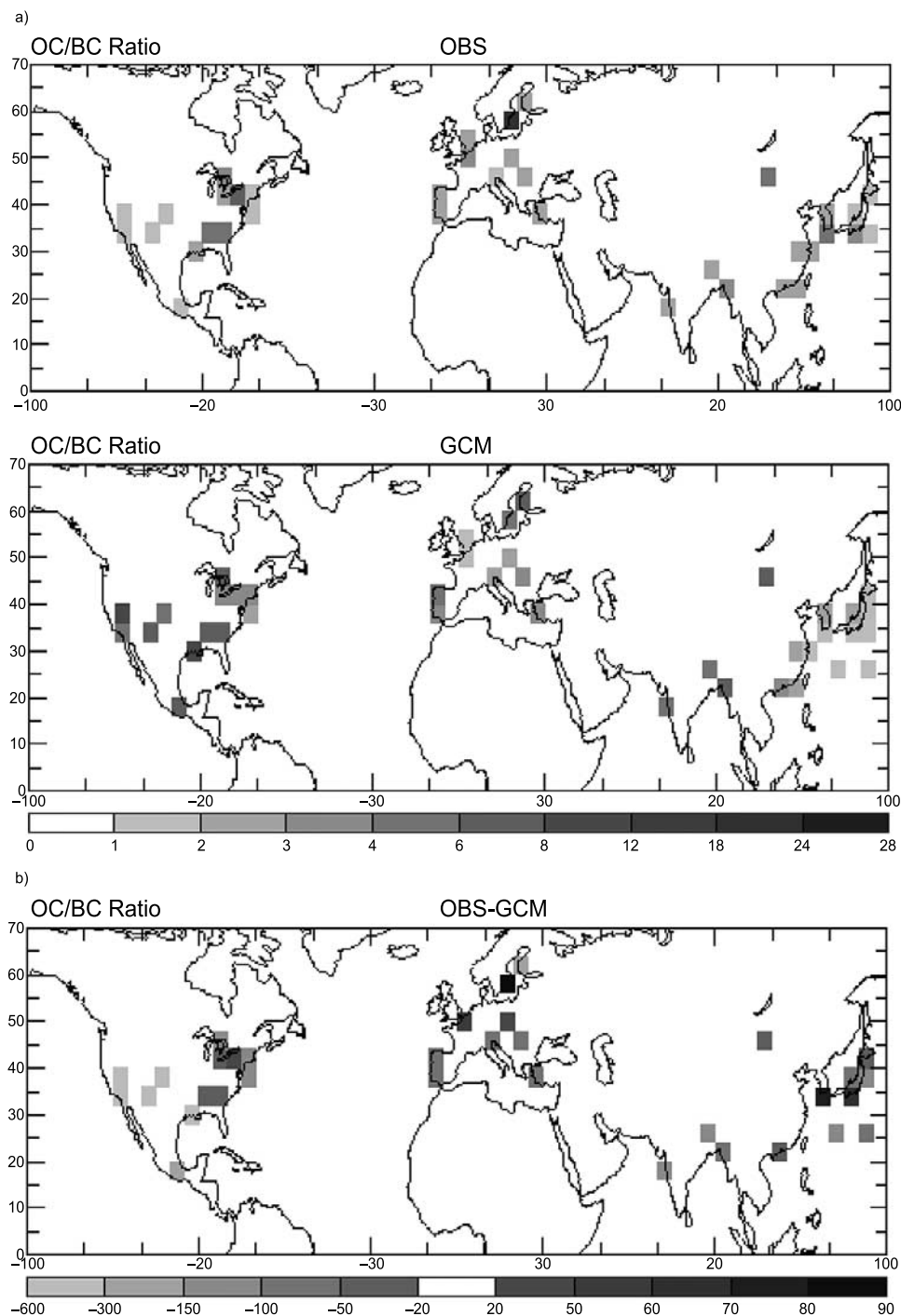
Figure 5. (a) Observed OC/BC ratios and model (GISS GCM) predicted OC/BC ratios (based on annual averages) at the select locations where measurements were available; (b) Differences (in percent) between measurements and model predicted OC/BC ratios. See color version of this figure at back of this issue.

Mediterranean with closer agreement (within 20%) mainly over China (Figure 5b).

4. Conclusions

[32] Our analysis of measured ambient OC/BC mass concentration ratios demonstrates that the neglect of a positive sampling artifact for OC results in a large overes-

timation of the OC/BC ratios, especially at low BC concentration. A survey of relevant publications (Table 1) shows that most data need to be corrected for this artifact. We have developed criteria to select those uncorrected OC/BC ratios that can be used as reasonable approximations for correct data. These criteria are defined using the results of studies in which both corrected and uncorrected ratios were determined.

[33] After artifact correction, the OC/BC ratios exhibit remarkable constancy for specific regions. Overall the ratios range from about 1.3 in Japan to about 2.4 at other locations. The OC/BC ratio shows no significant seasonal variability and appears to be insensitive to regional fuel and technology mix.

[34] Comparison of our empirical regional ratios with those calculated from emission inventories for both fossil and biomass sources [*Bond et al.*, 2004] show good agreement for China and Europe. Our ratios, however, are about a factor of 2 lower than the values calculated from OC and BC emission inventories for all other regions.

[35] The generally reduced values of OC/BC that we find after correction for the positive OC artifact has relevance to the climate effect of anthropogenic soot aerosols. It has been suggested [*Hansen et al.*, 2000; *Jacobson*, 2001] that reduction of anthropogenic BC emissions would help slow global warming. *Penner et al.* [2003], on the other hand, have pointed out that the cooling effect of OC that inevitably accompanies BC, together with the indirect effects of both BC and OC on cloud properties, make it uncertain whether the net effect of soot (BC + OC) emissions is warming or cooling. *Hansen et al.* [2005] calculate that the net effect of fossil fuel soot is warming, while biomass burning produces global cooling. The effect of reduced estimates for the OC/BC ratio, given in this paper, is to tilt the calculations for the effect of soot on climate more toward warming. The most effective targets for soot emission reductions would be those with especially low values of OC/BC, such as vehicles using diesel fuel. Reduction of aerosol emissions from such sources are most likely to reduce global warming as well improve public health [*Schneider and Hill*, 2005].

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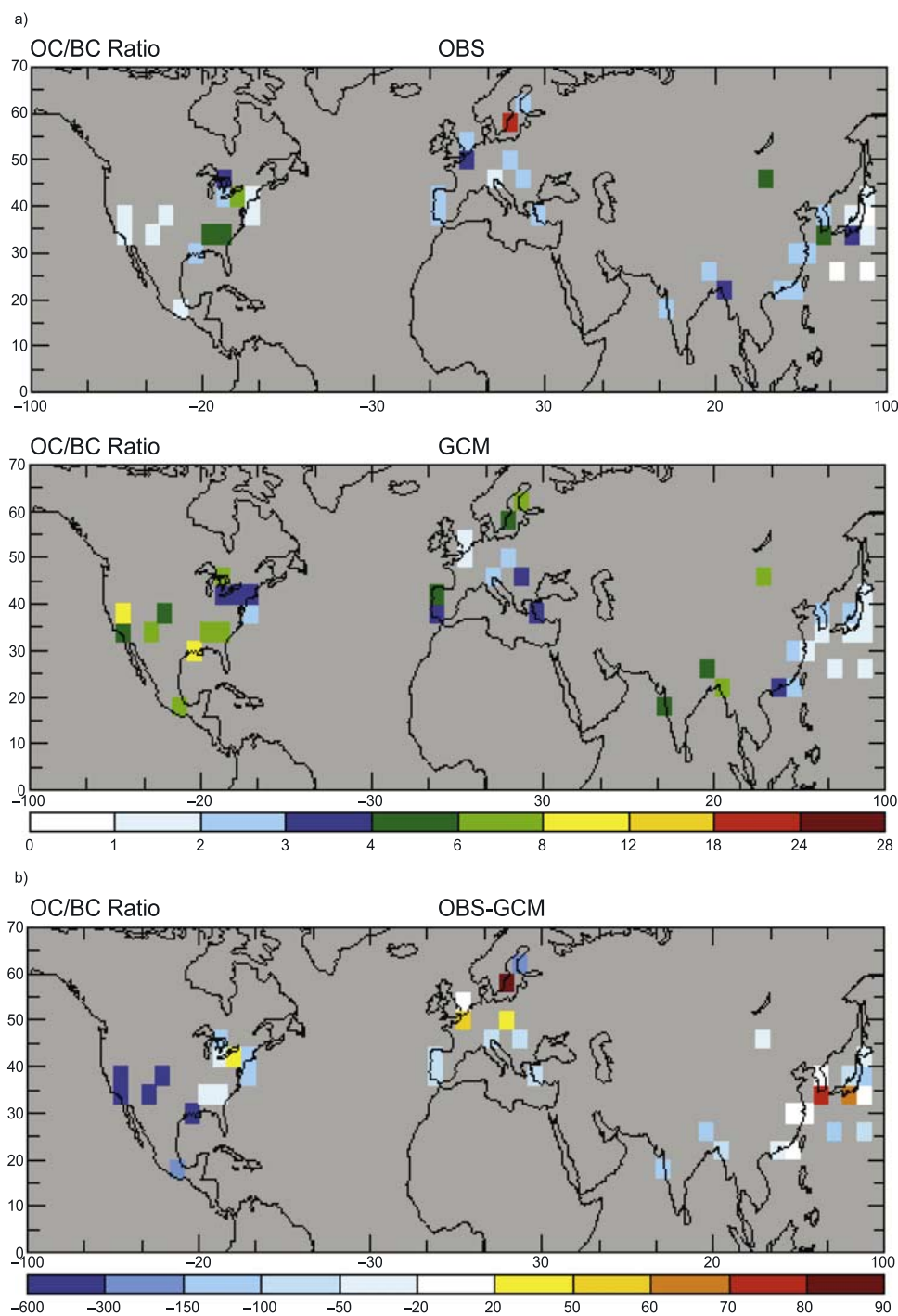
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Figure 5. (a) Observed OC/BC ratios and model (GISS GCM) predicted OC/BC ratios (based on annual averages) at the select locations where measurements were available; (b) Differences (in percent) between measurements and model predicted OC/BC ratios.